

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property
Organization
International Bureau



(43) International Publication Date
29 December 2004 (29.12.2004)

PCT

(10) International Publication Number
WO 2004/113237 A1

(51) International Patent Classification⁷: C02F 1/50, 11/00

(21) International Application Number:
PCT/GB2004/002660

(22) International Filing Date: 21 June 2004 (21.06.2004)

(25) Filing Language: English

(26) Publication Language: English

(30) Priority Data:
0314363.3 20 June 2003 (20.06.2003) GB

(71) Applicants (for all designated States except US): **RHODIA CONSUMER SPECIALTIES LIMITED** [GB/GB]; Oak House, Reeds Crescent, Watford, Hertfordshire WD24 4QP (GB). **THAMES WATER UTILITIES LIMITED** [GB/GB]; Clearwater Court, Vastern Road, Reading, Berks RG1 8DB (GB).

(72) Inventors; and

(75) Inventors/Applicants (for US only): **EDMUNDS, Stephanie** [GB/GB]; 18 St. Annes Road, Willenhall, West Midlands WV13 1ED (GB). **GILBERT, Paul,**

Douglas [GB/GB]; Alexandra, Sunray Avenue, Whitstable, Kent CT5 4EJ (GB). **TALBOT, Robert, Eric** [GB/GB]; 3 Meriden close, Cannock, Staffordshire WS11 1QG (GB). **ASAADI, Manocher** [GB/GB]; 60 Belleisle, Purley-on-Thames, Reading, Berks. RG8 8AP (GB). **WINTER, Peter** [DE/GB]; 127 St Peters Road, Reading, Berkshire RG6 1PG (GB).

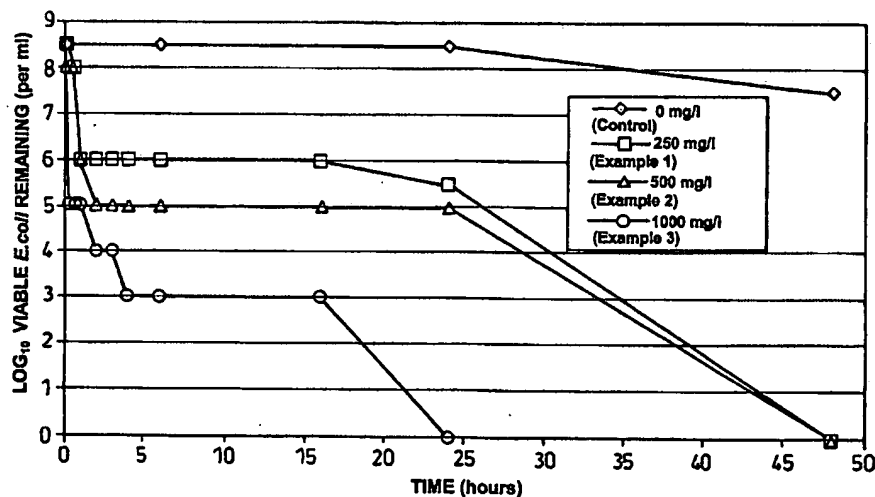
(74) Agent: **BARKER BRETTELL**; 138 Hagley Road, Edgbaston, Birmingham B16 9PW (GB).

(81) Designated States (unless otherwise indicated, for every kind of national protection available): AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BW, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DE, DK, DM, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NA, NI, NO, NZ, OM, PG, PH, PL, PT, RO, RU, SC, SD, SE, SG, SK, SL, SY, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, YU, ZA, ZM, ZW.

(84) Designated States (unless otherwise indicated, for every kind of regional protection available): ARIPO (BW, GH, GM, KE, LS, MW, MZ, NA, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM),

[Continued on next page]

(54) Title: TREATMENT OF SEWAGE SLUDGE



(57) Abstract: The present invention provides a method of reducing the pathogen content of sewage sludge comprising (a) adding to the sludge an effective amount of a phosphorous-containing compound and (b) keeping the phosphorous-containing compound in contact with the sludge for a sufficient time to reduce the amount of pathogens present in the sludge by an amount equivalent to a logarithmic reduction of 2 or more.



European (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HU, IE, IT, LU, MC, NL, PL, PT, RO, SE, SI, SK, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).

— *before the expiration of the time limit for amending the claims and to be republished in the event of receipt of amendments*

Published:

— *with international search report*

For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.

TREATMENT OF SEWAGE SLUDGE

This invention relates to a method of treating sewage sludge and to a sludge treated by the aforesaid method.

5

The treatment of raw sewage generally includes a filtration stage (in which large solids and grit are removed) followed by a stage in which the aqueous phase is subjected to aerobic bacterial action to remove biodegradable substances. This latter stage involves "activated sludge" which is essentially a concentrated bacterial mass. Biodegradable substances need to be removed prior to the discharge of the aqueous phase into watercourses, e.g. rivers, otherwise the bacterial degradation of such substances in the river would consume dissolved oxygen resulting in fish deaths, odours and general degradation of the environment. During the degradation of the biodegradable substances, growth and multiplication of the bacteria occur, resulting in the accumulation of bacterial sludge requiring disposal.

Optionally, the excess sludge may be "digested" under anaerobic conditions where, essentially, the bacteria re-equilibrate under the new conditions to produce methane and reduce the biomass but, ultimately, there remains an irreducible mass of excess sludge which requires disposal. There are a number of methods of disposal, such as landfill and disposal at sea, both of which are disfavoured for environmental reasons. Alternatively, the excess sludge may be incinerated (expensive) or spread on to agricultural land and, in the latter case, the sludge can be used as a fertiliser/soil conditioner, which is a benefit.

Unfortunately, such sludge can contain significant concentrations of pathogens and, if so, the sludge requires disinfection to reduce to an acceptable environmental and sanitary level any pathogenic organisms

present, before the disinfected sludge is spread to land. An indicator organism, used to quantify the pathogenic risk, is *E. coli*. For compliance with UK statutory provisions, for conventional treated sludge the level of *E. coli* in the sludge must be reduced by 99% (i.e. a
5 logarithmic reduction of 2) and the maximum acceptable level of *E. coli* in the treated sewage sludge is 10^5 per gram of dry sludge (gds). For enhanced treated sludge in the UK there should be no *Salmonella spp* present and the level of *E. coli* must be reduced by at least 99.9999% (i.e. a logarithmic reduction of 6). The maximum acceptable level of *E.*
10 *coli* in the enhanced treated sewage sludge is 10^3 per gram of dry sludge. Similar statutory requirements are expected to be adopted across Europe and in the USA in the future.

Bacterial reduction may be accomplished in a variety of ways including
15 lime treatment (messy, requires significant capital investment and poses severe handling problems) heat treatment (very expensive) or merely leaving the sludge in storage till the bacterial level falls within the required limit. For the latter situation, the very large volumes of sludge involved at most sewage treatment works cannot usually be stored for the
20 requisite time due to insufficient storage capacity. Installing sufficient capacity is either impractical due to space considerations or involves large capital expenditure.

In theory, an alternative method of reducing the bacterial content of the
25 sludge would be to apply a disinfectant. However, disinfectants evaluated hitherto have been found to take relatively long periods to reduce the bacterial content to an acceptable level, thus creating storage demands beyond the resources of most sewage-treatment works.

30 We have found that the use of a phosphorus-containing compound (especially a phosphonium salt) on sewage sludge can bring about a

reduction in the pathogen content of the sludge equivalent to a logarithmic decrease of at least 2.

Accordingly, the present invention provides a method of treating sewage
5 sludge to reduce the pathogen content of said sludge, the method comprising the steps of:

- (a) adding to the sludge an effective amount of a phosphorus-containing compound; and
- 10 (b) keeping the phosphorus-containing compound in contact with the sludge for sufficient time to reduce the amount of pathogens present in the sludge by an amount equivalent to a logarithmic reduction of 2 or more.

15

In one embodiment the log reduction of 2 or more is achieved over a 24-hour period.

20

Preferably, the phosphorus-containing compound is kept in contact with the sludge for sufficient time to reduce the amount of pathogens present in the sludge by a log reduction of 3 or more and more preferably 4 or more.

25

The pathogens may be bacteria.

Preferably, the sludge has undergone anaerobic digestion, a process known to those skilled in the art, prior to step (a).

30

Preferably, the phosphorus-containing compound is a phosphonium compound, especially a tetrakis(hydroxyorgano)phosphonium salt or compound of formula (I)



wherein:

5 n is the valency of X;

R' and R'', which may be the same or different, are selected from an alkyl, hydroxyalkyl, alkenyl or aryl moiety and X is an anion.

R' and R'' are preferably between 1 and 20 carbon atoms in length.

10

X is preferably selected from the group consisting of chloride, sulphate, phosphate, acetate, oxalate and bromide.

Most preferably, the phosphonium compound is tetrakis(hydroxymethyl)
15 phosphonium sulphate.

Alternatively, the phosphonium compound may be, for example, a tetrakis(hydroxymethyl) phosphonium chloride, tetrakis(hydroxymethyl) phosphonium bromide, tetrakis(hydroxymethyl)phosphonium phosphate,
20 tetrakis(hydroxymethyl)phosphonium acetate or tetrakis(hydroxymethyl)phosphonium oxalate.

Alternatively, the phosphorus-containing compound may be an alkyl-substituted phosphine, e.g. tris(hydroxymethyl) phosphine as shown in
25 formula (II):



wherein:

each R, which may be the same or different, is selected from a alkyl,
30 hydroxyalkyl, alkenyl or aryl moiety.

The amount of phosphorus-containing compound to be added to the sludge in step (a) of the method of the present invention is suitably up to 10000mg/l, preferably 100-2500mg/l, and especially 200-1000mg/l.

- 5 Alternatively, the amount of phosphorus-containing compound to be added to the sludge may be expressed relative to dry solids weight. Suitably, the amount to be added is up to about 30% by weight of dry solids. Preferably, the amount of phosphorus-containing compound to be added may be from 0.1 to 20%, for example, 0.1 to 10%, such as 0.2 to
10 5% or 0.4 to 2% by weight of dry solids.

Step (b) of the method of the present invention may be carried out over a period of from 1 second to 14 days. For example, from 6 to 24 hours, from 1 to 6 hours, from 1 to 60 minutes, from 1 to 60 seconds or from 1
15 to 15 seconds.

The rate of addition of the phosphorus-containing compound and the rate of mixing are important in maximising the efficacy of the process. To maximise efficacy, both should be as short as practically possible and
20 contact time should be maximised. In processes involving natural gravity settling of the sewage sludge step (b) is preferably 6 to 24 hours. In processes where the treated sludge is, optionally, dewatered by, e.g. centrifuge or filter press, to produce 'sludge cake', step (b) is preferably carried out in 15 seconds to 24 hours. 'Sludge cake' has substantially
25 higher solids content than liquid sludge. Dewatering aids such as polydiallyl-dimethyl ammonium chlorides, polyamines, cationised polyacrylamides and anionic polyacrylamides may be utilised in the production of 'sludge cake'.

- 30 The pathogens present in the sludge are suitably selected from the group consisting of:

- bacteria, including *Escherichia coli*, *Salmonella spp.*, *Shigella spp.*, *Vibrio cholerae*, *Bacillus cereus*, *Listeria monocytogenes*, *Campylobacter spp* and *Yersinia pestis*;
- 5 • viruses, including rotaviruses, calciviruses, group F adenoviruses and astroviruses;
- protozoans, including *Entamoeba spp.*, *Giardia spp.*, *Balantidium coli* and *Cryptosporidium spp.*; and
- 10 • helminths and their eggs, including nematodes, for example, *Ascaris lumbricoides* (roundworm), *Trichuris trichiura* (whipworm), *Ancylostoma duodenale* (hookworm), *Strongyloides stercoralis* (threadworm); trematodes, for example, *Schistosoma spp.*; and cestodes, for example, *Taenia saginata* (beef tapeworm) and *Taenia solum* (pork tapeworm).

15

Preferably the method according to the present invention provides from a two to six log reduction of the pathogens present in the sludge.

- 20 A two-log reduction is defined by 99% of the pathogens present in the sludge being eliminated. Sludge treated in this way is termed 'conventionally treated sludge'. A six-log reduction is defined by 99.9999% of the pathogens present in the sludge being eliminated. Sludge treated in this way is termed 'enhanced treated sludge'.

25

The present invention further provides a sewage sludge that has been treated according to the method described hereinabove.

- 30 The present invention will be illustrated by way of the following Examples.

In the Examples, the phosphorus-containing compound used to treat sewage sludge was 75% w/w tetrakis(hydroxymethyl) phosphonium sulphate, available from Rhodia Consumer Specialties Limited. For the purposes of this patent specification, the product will be subsequently referred to as "Phosphonium Salt".

As a comparison, sewage sludges were treated with a conventional disinfectant compound, dibromo-nitrilo-propionamide (DBNPA).

In each Example, the bacterium being observed was *E. coli*.

1.1 METHODOLOGY

The methodology adopted to evaluate biocide performance was by Quantitative Suspension Test (QST) using sterile anaerobic digester sludge as the QST medium, back-inoculated with *E. coli* cultures previously isolated from the sludge. In this way, a consistent chemical environment (sterile sludge) could be used in conjunction with a defined bacterial challenge. This enables the provision of consistency between tests.

1.2 MICROBIOLOGICAL EVALUATIONS

Sterile sludge was prepared from raw sludge samples by autoclaving at 121°C for 20 minutes. The *E. coli* strains used in QST had been isolated from raw sludge samples.

QST were performed as follows:

- Sterile sludge (19ml) was dispensed into sterile, screw-cap, plastics universal bottles of nominal 30ml capacity.

- 5 • To each sample was added 0.5ml of a washed cell suspension of *E. coli* prepared from a 16-hour culture incubated at 44°C in Tryptose Soy Broth, which had been centrifuged (14500 rpm for 10 min.) and re-suspended in sterile phosphate buffer (0.2M at pH 7.2). An inoculum of 0.5ml was sufficient to provide a final cell concentration of about 10^8 per ml in 20 ml of QST medium.
- 10 • Fresh stock solutions of the candidate treatment chemicals were prepared in sterile phosphate buffer (0.2M at pH 7.2) at concentrations such that when 0.5ml was added to the QST medium (final volume 20ml) the desired final concentration of biocide was achieved.
- 15 • The QST medium was mixed thoroughly and held at 22°C for the duration of the test.
- 20 • At intervals during the test, the sludge was well mixed and samples (1.0ml) were removed from the QST medium and inoculated into the first tube of a dilution series containing MacConkey broth supplemented with sodium thiosulphate (5.0 g/l), to inactivate any residual biocide carried into the dilution series. This was carried out in duplicate.
- 25 • The remainder of the serial dilution (10 fold steps) was carried out in MacConkey Broth alone and tubes incubated at 44°C for 16 hours. The end point was scored as the highest dilution in the series to show a change in colour from purple to yellow and to have developed turbidity.

MacConkey Broth was selected as this medium contains the pH indicator Bromocresol Purple that changes from purple to yellow as the medium becomes acidic. This is a useful indirect indicator of microbial growth (organic acid production) where this cannot be scored by the development of turbidity in an initially clear medium. Because the sludge contains suspended solids the first 2 tubes of the dilution series instantaneously develop turbidity on the addition of the sludge. This precludes using turbidity alone as an indicator of microbial growth.

10 The biocides used in the evaluations are shown in the Table below.

BIOCIDE TYPE	ACTIVE INGREDIENT (ai)	PERCENT ai
Phosphonium Salt	THPS	75
DBNPA	DBNPA	98

EXAMPLES 1 to 3

15 The performance of Phosphonium Salt in the concentration range 250 to 1000mg/l is illustrated in Figure 1 of the accompanying drawings. Concentrations of 250 and 500 mg/l gave similar results with a fairly flat time/kill response over the first 6 hours contact time, followed by a reduction in numbers to a total kill within 48 hours.

20

By contrast, the time/kill response at 1000mg/l was much faster. The time/kill response over the first 6 hours contact time was more progressive and total kill was achieved within 24 hours.

25 For comparison, the *E. coli* levels in untreated sludge slowly decrease naturally, over a time period as shown in figure 2. Even starting at the low *E. coli* level of 10^4 cfu/gds it took 6 days to achieve total kill. Starting at the higher level of $10^{8.5}$ cfu/gds, the level had only reduced to

10

10⁴ cfu/gds after 8days. The benefit of phosphonium salt treatment (figure 1) is therefore effectively displayed.

EXAMPLE 4

5

The performance of Phosphonium Salt compared to that of DBNPA, is shown in Figure 3 of the accompanying drawings. Both biocides were tested at an equal active-ingredient concentration of 500mg/l. DBNPA shows surprisingly poor anti-microbial performance, achieving only a 2.5
10 log reduction in numbers after 48 hours.

The foregoing Examples demonstrate the following characteristics of the present invention:

- 15 (a) Increasing the Phosphonium Salt concentration used in treatment from 500 to 1000mg/l gives a significant improvement in performance.
- (b) In all of the treatments evaluated total kill was achieved.
- 20 (c) When compared with the performance of DBNPA, the performance of Phosphonium Salt was superior.

25

30

CLAIMS

1. A method of treating sewage sludge to reduce the pathogen content of said sludge, the method comprising the steps of:
 - 5 (c) adding to the sludge an effective amount of a phosphorus-containing compound; and
 - 10 (d) keeping the phosphorus-containing compound in contact with the sludge for sufficient time to reduce the amount of pathogens present in the sludge by an amount equivalent to a logarithmic reduction of 2 or more.
- 15 2. A method as claimed in claim 1 in which the log reduction of 2 or more is achieved over a 24-hour period.
- 20 3. A method as claimed in claim 1 in which the phosphorus-containing compound is kept in contact with the sludge for sufficient time to reduce the amount of pathogens present in the sludge by a log reduction of 3 or more.
- 25 4. A method as claimed in claim 3 in which the phosphorus-containing compound is kept in contact with the sludge for sufficient time to reduce the amount of pathogens present in the sludge by a log reduction of 4 or more.
5. A method as claimed in any one of the preceding claims in which the pathogens are bacteria.
- 30 6. A method as claimed in any one of the preceding claims in which the sludge has undergone anaerobic digestion prior to step (a).

7. A method as claimed in any one of the preceding claims in which the phosphorus-containing compound is a phosphonium compound.

8. A method as claimed in claim 7 in which the phosphonium
5 compound is a tetrakis(hydroxyorgano)phosphonium salt or compound of formula (I)



10 wherein:

n is the valency of X;

R' and R'', which may be the same or different, are selected from an alkyl, hydroxyalkyl, alkenyl or aryl moiety and X is an anion.

15 9. A method as claimed in claim 8 in which R' and R'' are between 1 and 20 carbon atoms in length.

10. A method as claimed in claim 8 or claim 9 in which X is selected from the group consisting of chloride, sulphate, phosphate, acetate,
20 oxalate and bromide.

11. A method as claimed in any one of claims 8 to 10 in which the phosphonium compound is tetrakis(hydroxymethyl) phosphonium sulphate.

25

12. A method as claimed in any one of claims 8 to 10 in which the phosphonium compound is selected from tetrakis(hydroxymethyl) phosphonium chloride, tetrakis(hydroxymethyl)phosphonium bromide, tetrakis(hydroxymethyl)phosphonium phosphate, tetrakis (hydroxymethyl)
30 phosphonium acetate or tetrakis(hydroxymethyl)phosphonium oxalate.

13

13. A method as claimed in any one of claims 1 to 6 in which the phosphorus-containing compound is an alkyl-substituted phosphine as shown in formula (II):



wherein:

each R, which may be the same or different, is selected from a alkyl, hydroxyalkyl, alkenyl or aryl moiety.

10 14. A method as claimed in any one of the preceding claims in which the amount of phosphorus-containing compound to be added to the sludge in step (a) of the method of the present invention is up to 10000mg/l.

15 15. A method as claimed in claim 14 in which the amount of phosphorus-containing compound to be added to the sludge in step (a) of the method of the present invention is 100-2500mg/l.

20 16. A method as claimed in claim 15 in which the amount of phosphorus-containing compound to be added to the sludge in step (a) of the method of the present invention is 200-1000mg/l.

25 17. A method as claimed in any one of claims 1 to 13 in which the amount of phosphorus-containing compound to be added to the sludge is expressed relative to dry solids weight and the amount to be added is up to about 30% by weight of dry solids.

18. A method as claimed in claim 17 in which the amount of phosphorus-containing compound to be added is from 0.1 to 20% by weight of dry solids.

30

19. A method as claimed in claim 18 in which the amount of phosphorus-containing compound to be added is from 0.1 to 10% by weight of dry solids.
- 5 20. A method as claimed in claim 17 in which the amount of phosphorus-containing compound to be added is from 0.2 to 5% by weight of dry solids.
- 10 21. A method as claimed in claim 17 in which the amount of phosphorus-containing compound to be added is from 0.4 to 2% by weight of dry solids.
22. A method as claimed in any one of the preceding claims in which step (b) of the method of the present invention is carried out over a period
15 of from 1 second to 14 days.
23. A method as claimed in claim 22 in which step (b) of the method of the present invention is carried out over a period of from 6 to 24 hours.
- 20 24. A method as claimed in claim 22 in which step (b) of the method of the present invention may be carried out over a period of from 15 seconds to 24 hours.
- 25 25. A method as claimed in any one claims 1 to 4 and claims 6 to 24 in which the pathogens present in the sludge are selected from the group consisting of bacteria, viruses, protozoans and helminths.
- 30 26. A method as claimed in claim 5 and 25 in which the bacteria are selected from the group consisting of *Escherichia coli*, *Salmonella spp.*, *Shigella spp.*, *Vibrio cholerae*, *Bacillus cereus*, *Listeria monocytogenes*, *Campylobacter spp.* and *Yersinia pestis*.

27. A method as claimed in claim 25 in which the viruses are selected from the group consisting of rotaviruses, calciviruses, group F adenoviruses and astroviruses.

5

28. A method as claimed in claim 25 in which the protozoans are selected from the group consisting of *Entamoeba spp.*, *Giardia spp.*, *Balantidium coli* and *Cryptosporidium spp.*

10 29. A method as claimed in claim 25 in which the helminths are selected from the group consisting of *Ascaris lumbricoides* (roundworm), *Trichuris trichiura* (whipworm), *Ancylostoma duodenale* (hookworm), *Strongyloides stercoralis* (threadworm), *Schistosoma spp.*, *Taenia saginata* (beef tapeworm), *Taenia solum* (pork tapeworm) and their eggs.

15

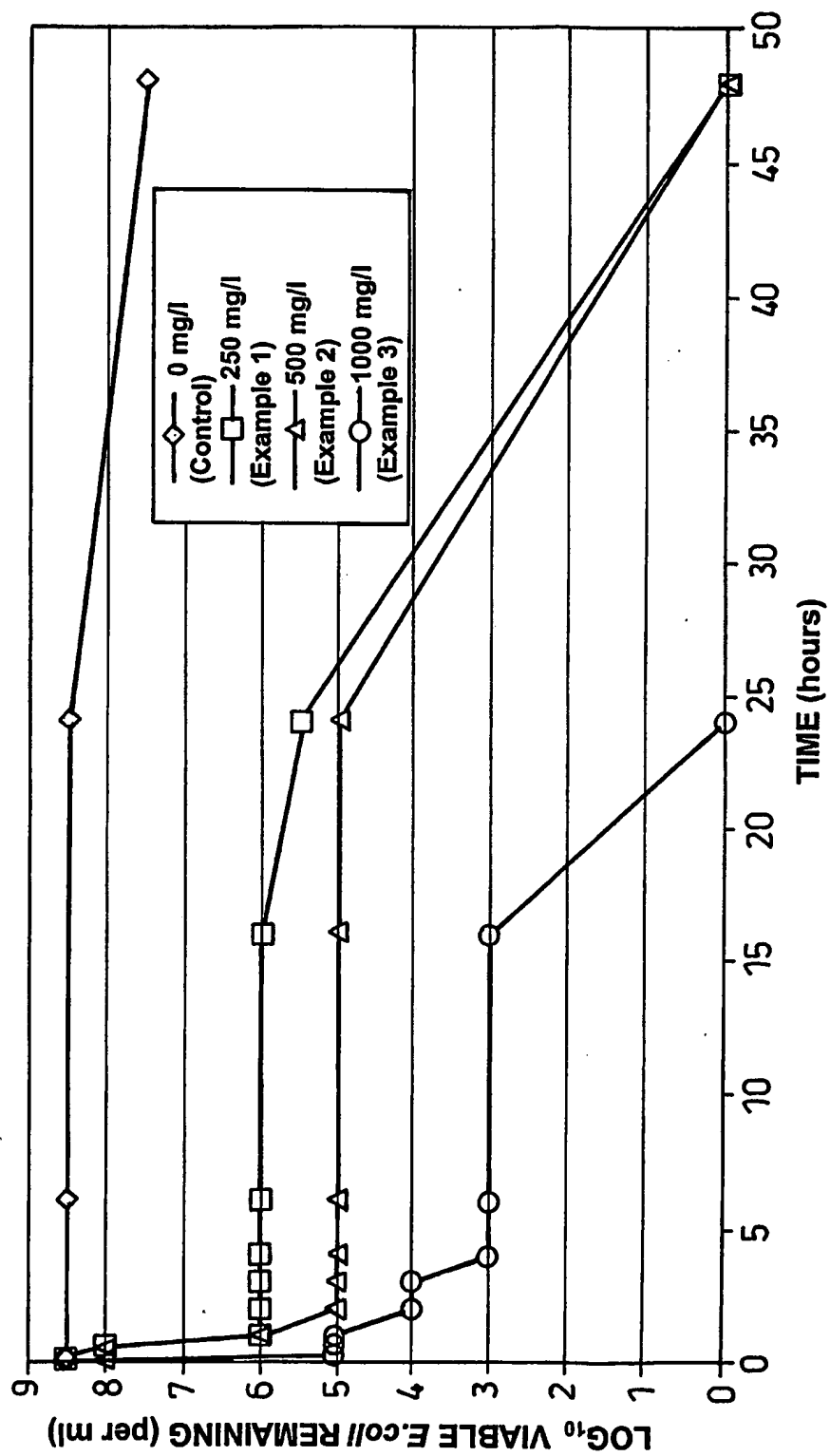
30. A sewage sludge that has been treated according to the method as claimed in any one of claims 1 to 29.

20 31. A method of treating sewage sludge substantially as described herein with reference to the accompanying examples and figures.

32. A treated sludge substantially as described herein with reference to the accompanying examples and figures.

25

1/3

*Fig. 1*

2/3

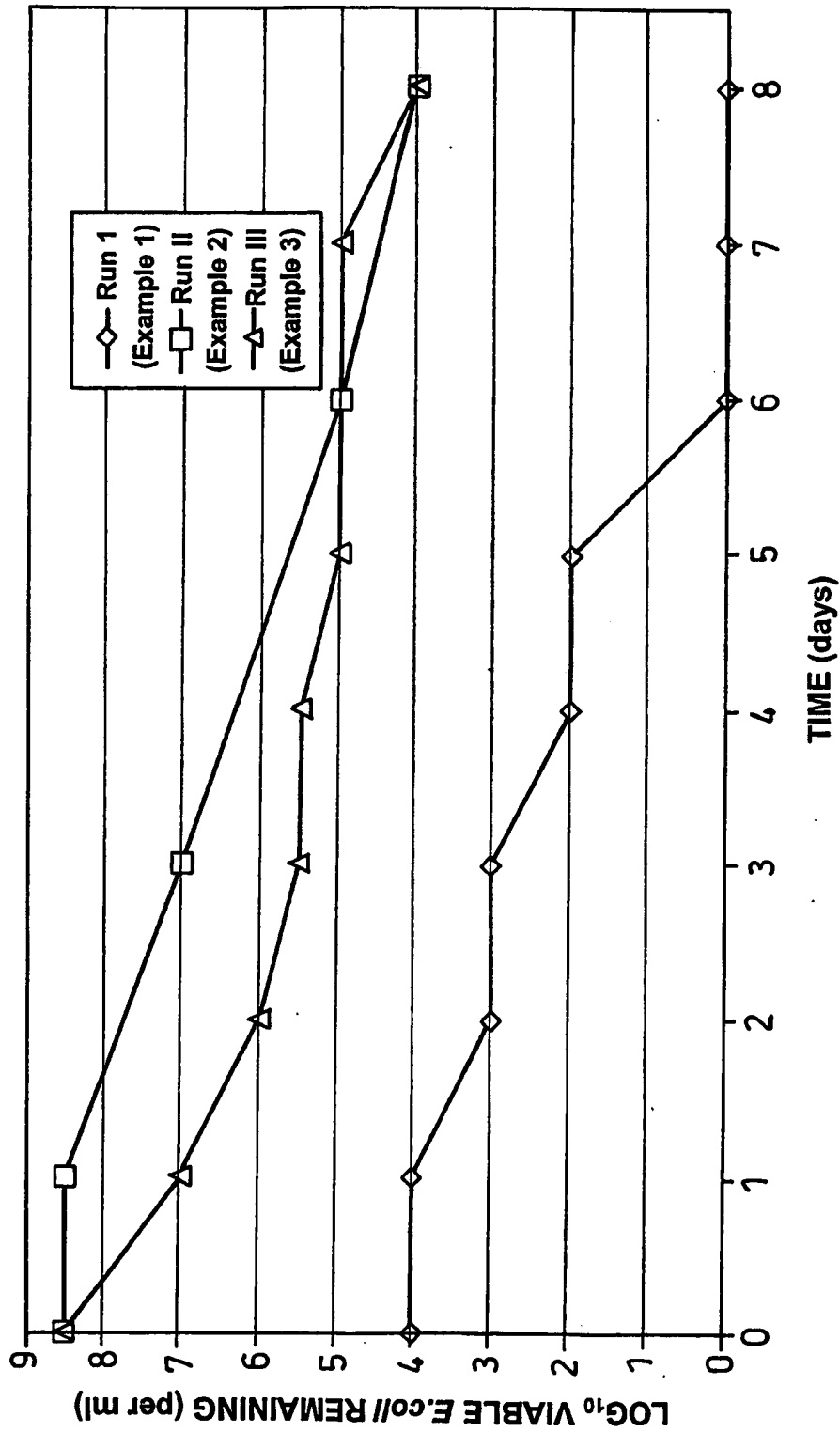
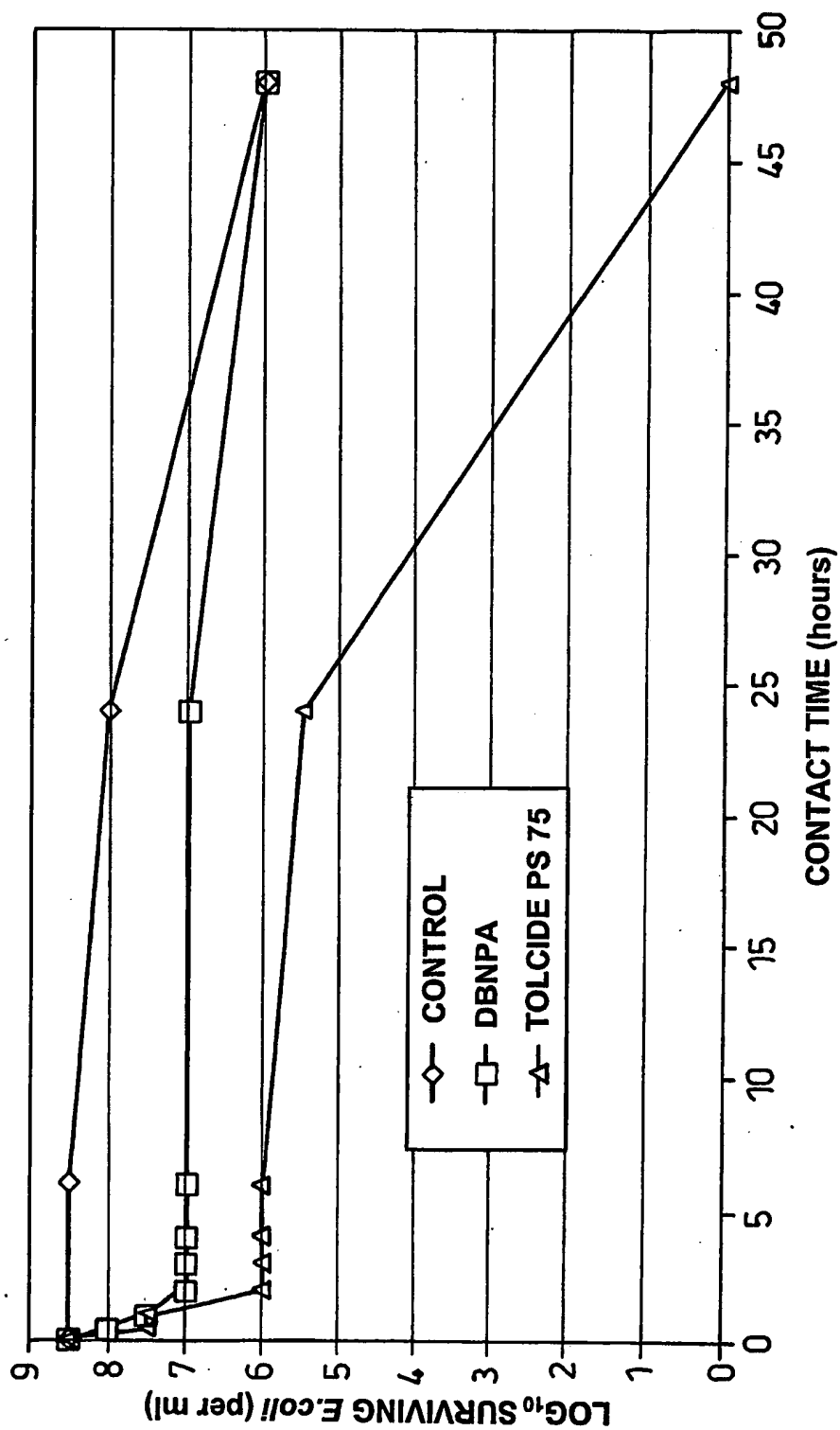


Fig. 2

3/3

*Fig. 3*

INTERNATIONAL SEARCH REPORT

International Application No.

GB2004/002660

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 C02F1/50 C02F11/00

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 C02F

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, PAJ, WPI Data, COMPENDEX, BIOSIS, EMBASE

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 422 015 A (ANGELL EDWIN C ET AL) 6 June 1995 (1995-06-06) claims 1,2	1,22,24, 25,30
X	FR 2 794 118 A (RHONE POULENC CHIMIE) 1 December 2000 (2000-12-01) page 2, line 1 - line 5; claims 1,3,4,14 page 3, line 27 - line 30	1,6,7,30
X	US 5 965 033 A (DEL GROSSO BIRGIT ET AL) 12 October 1999 (1999-10-12) column 3, line 28 - line 30; claims 1,6 column 4, line 7 - line 30	1,5,25, 30
	----- -/--	

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents:

- *A* document defining the general state of the art which is not considered to be of particular relevance
- *E* earlier document but published on or after the international filing date
- *L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- *O* document referring to an oral disclosure, use, exhibition or other means
- *P* document published prior to the international filing date but later than the priority date claimed

- *T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- *X* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- *Y* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
- *Z* document member of the same patent family

Date of the actual completion of the international search

27 October 2004

Date of mailing of the international search report

05/11/2004

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2
NL - 2280 HV Rijswijk
Tel (+31-70) 340-2040, Tx. 31 651 apo nl,
Fax: (+31-70) 340-3016

Authorized officer

Gonzalez Arias, M

INTERNATIONAL SEARCH REPORT

International Application No

/GB2004/002660

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	<p>PATENT ABSTRACTS OF JAPAN vol. 2003, no. 02, 5 February 2003 (2003-02-05) & JP 2002 308713 A (AQUAS CORP; K I CHEMICAL INDUSTRY CO LTD), 23 October 2002 (2002-10-23) abstract</p>	
A	<p>EP 0 215 562 A (ALBRIGHT & WILSON) 25 March 1987 (1987-03-25) column 5, line 7 column 12, line 10 - line 54</p>	<p>1,7,13, 25</p>

INTERNATIONAL SEARCH REPORT

International application No.
PCT/GB2004/002660

Box II Observations where certain claims were found unsearchable (Continuation of item 2 of first sheet)

This International Search Report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☒ Claims Nos.: 31, 32
because they relate to parts of the International Application that do not comply with the prescribed requirements to such an extent that no meaningful International Search can be carried out, specifically:
see FURTHER INFORMATION sheet PCT/ISA/210
3. ☐ Claims Nos.:
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

Box III Observations where unity of invention is lacking (Continuation of item 3 of first sheet)

This International Searching Authority found multiple inventions in this International application, as follows:

1. ☐ As all required additional search fees were timely paid by the applicant, this International Search Report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this International Search Report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☐ No required additional search fees were timely paid by the applicant. Consequently, this International Search Report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest.
- ☐ No protest accompanied the payment of additional search fees.

FURTHER INFORMATION CONTINUED FROM PCT/ISA/ 210

Continuation of Box II.2

Claims Nos.: 31,32

lack of technical features. Rule 6.2(a) PCT

The applicant's attention is drawn to the fact that claims relating to inventions in respect of which no international search report has been established need not be the subject of an international preliminary examination (Rule 66.1(e) PCT). The applicant is advised that the EPO policy when acting as an International Preliminary Examining Authority is normally not to carry out a preliminary examination on matter which has not been searched. This is the case irrespective of whether or not the claims are amended following receipt of the search report or during any Chapter II procedure. If the application proceeds into the regional phase before the EPO, the applicant is reminded that a search may be carried out during examination before the EPO (see EPO Guideline C-VI, 8.5), should the problems which led to the Article 17(2) declaration be overcome.

INTERNATIONAL SEARCH REPORT

International Application No

PC/GB2004/002660

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
US 5422015	A	06-06-1995	US 5482528 A	09-01-1996
FR 2794118	A	01-12-2000	FR 2794118 A1	01-12-2000
			AU 5227900 A	18-12-2000
			EP 1196356 A1	17-04-2002
			WO 0073219 A1	07-12-2000
US 5965033	A	12-10-1999	DE 19531241 A1	27-02-1997
			AT 184580 T	15-10-1999
			BR 9610434 A	15-06-1999
			CA 2229541 A1	06-03-1997
			DE 59603100 D1	21-10-1999
			WO 9708100 A1	06-03-1997
			EP 0851842 A1	08-07-1998
			ES 2138371 T3	01-01-2000
			GR 3031948 T3	31-03-2000
			JP 11514283 T	07-12-1999
			DK 851842 T3	03-04-2000
JP 2002308713	A	23-10-2002	NONE	
EP 0215562	A	25-03-1987	AT 56585 T	15-10-1990
			AU 597894 B2	14-06-1990
			AU 6087686 A	12-02-1987
			DE 3674308 D1	25-10-1990
			EP 0215562 A1	25-03-1987
			FI 863214 A ,B,	07-02-1987
			GB 2178960 A ,B	25-02-1987
			IN 166861 A1	28-07-1990
			JP 2004977 C	11-01-1996
			JP 6099255 B	07-12-1994
			JP 62042908 A	24-02-1987
			KR 9203210 B1	24-04-1992
			MX 173125 B	02-02-1994
			NO 863154 A ,B,	09-02-1987
			US 5741757 A	21-04-1998
			ZA 8605846 A	25-03-1987
			CA 1272559 A1	14-08-1990